U.S. Application No.: 10/074,014

Attorney Docket No.: Q68466

AMENDMENTS TO THE CLAIMS

This listing of claims will replace all prior versions and listings of claims in the application:

LISTING OF CLAIMS:

- 1-8. (canceled).
- 9. (withdrawn): A method of preparing an isothiocyanatoformic acid ester derivative represented by the following general formula (2):

General formula (2)

in which: R⁴ represents a substituted or non-substituted alkyl group, or a substituted or non-substituted aryl group; R⁵ represents a substituted or non-substituted alkyl group, a substituted or non-substituted aryl group, or a substituted or non-substituted heterocyclic group; and R⁶ represents a substituted or non-substituted alkyl group having at least 3 carbon atoms, or a substituted or non-substituted aryl group, the method comprising the steps of:

adding a chloroformic acid derivative represented by the following general formula (7) to an isothiocyanic acid salt represented by the following general formula (5) and a hydroxy derivative represented by the following general formula (6) for preparing an intermediate represented by the following general formula (8); and

preparing the isothiocyanatoformic acid ester derivative from the intermediate:

U.S. Application No.: 10/074,014

Attorney Docket No.: Q68466

ZNCS General formula (5) R⁶OH General formula (6)

O || CICOR⁴ O S || || R⁴OCNHCOR⁶

General formula (7)

General formula (8)

wherein, in the general formula (5), Z represents a sodium atom or a potassium atom.

- 10. (canceled).
- 11. (withdrawn): A method of preparing an isothiocyanatoformic acid ester derivative represented by the following general formula (2):

General formula (2)

in which: R⁴ represents a substituted or non-substituted alkyl group, or a substituted or non-substituted aryl group; R⁵ represents a substituted or non-substituted alkyl group, a substituted or non-substituted aryl group, or a substituted or non-substituted heterocyclic group;

U.S. Application No.: 10/074,014

Attorney Docket No.: Q68466

and R⁶ represents a substituted or non-substituted alkyl group having at least 3 carbon atoms, or a substituted or non-substituted aryl group, the method comprising the steps of:

preparing an intermediate represented by the following general formula (10); and reacting the intermediate with an alkylating agent represented by one of the following general formula (11) and the following general formula (12) for preparing the isothiocyanatoformic acid ester derivative:

$$\begin{pmatrix}
O \\
R^4OCN = C
\end{pmatrix}
C
S$$

$$N$$

$$N$$

$$N$$

$$N$$

$$N$$

$$N$$

$$N$$

$$N$$

General formula (10)

General formula (11)

wherein, in the general formula (10), M represents an alkali metal atom, an alkali earth metal atom, an aluminum atom or a magnesium atom, and, in the general formula (11), X represents a halogen atom or SO₃Ar, and Ar represents a substituted or non-substituted aryl group.

12. (currently amended): A method of preparing an isothiocyanatoformic acid ester derivative product representative by the following formula (2):

U.S. Application No.: 10/074,014 Attorney Docket No.: Q68466

Formula (2)

in which: R⁴ represents a substituted or non-substituted alkyl group, or a substituted or non-substituted aryl group; R⁵ represents a substituted or non-substituted alkyl group, a substituted or non-substituted aryl group, or a substituted or non-substituted heterocyclic group; and R⁶ represents a substituted or non-substituted alkyl group having at least 3 carbon atoms, or a substituted or non-substituted aryl group, the method comprising the steps of:

preparing a first intermediate represented by the following formula (8) by adding-a ehloroformic acid derivative represented by the following formula (7) to an isothiocyanic acid salt represented by the following formula (5), and a hydroxy derivative represented by the following formula (6), and further adding a chloroformic acid derivative represented by the following formula (7) for preparing a first intermediate represented by the following formula (8);

preparing a second intermediate represented by the following formula (10) from the first intermediate and a compound represented by the following formula (9); and

reacting the second intermediate with an alkylating agent represented by one of the following formulas (11) and (12) for preparing the isothiocyanatoformic acid ester-derivative product-from the second intermediate:

U.S. Application No.: 10/074,014

Formula (11)

Attorney Docket No.: Q68466

R⁶OH **ZNCS** Formula (6) Formula (5) Formula (8) Formula (7) $M(OH)_n$ Formula (10) Formula (9) $(R^5O)_2SO_2$ R^5X Formula (12)

wherein, in the formula (5), Z represents a sodium atom or a potassium atom, and, in the formulas (9) and (10), M represents an alkali metal atom, an alkali earth metal atom, an

aluminum atom or a magnesium atom, n represents an integer selected from 1 to 3, and, in the formula (11), X represents a halogen atom or SO₃Ar, and Ar represents a substituted or non-substituted aryl group, and

wherein the second intermediate represented by the formula (10) is obtained as crystals.

13. (withdrawn): A method of preparing an isothiocyanatoformic acid ester derivative represented by the following general formula (2):

General formula (2)

in which: R⁴ represents a substituted or non-substituted alkyl group, or a substituted or non-substituted aryl group; R⁵ represents a substituted or non-substituted alkyl group, a substituted or non-substituted aryl group, or a substituted or non-substituted heterocyclic group; and R⁶ represents a substituted or non-substituted alkyl group having at least 3 carbon atoms, or a substituted or non-substituted aryl group, the method comprising the steps of:

preparing a first intermediate represented by the following general formula (8);

preparing a second intermediate represented by the following general formula (10) from the first intermediate and a compound represented by the following general formula (9); and

reacting the second intermediate with an alkylating agent represented by one of the following general formula (11) and the following general formula (12) for preparing the isothiocyanatoformic acid ester derivative:

General formula (12)

 $(R^5O)_2SO_2$

wherein, in the general formulas (9) and (10), M represents an alkali metal atom, an alkali earth metal atom, an aluminum atom or a magnesium atom, and, in the general formula (11), X represents a halogen atom or SO₃Ar, and Ar represents a substituted or non-substituted aryl group.

14. (withdrawn): A method of preparing an isothiocyanatoformic acid ester derivative represented by the following general formula (2):

U.S. Application No.: 10/074,014 Attorney Docket No.: Q68466

General formula (2)

in which: R⁴ represents a substituted or non-substituted alkyl group, or a substituted or non-substituted aryl group; R⁵ represents a substituted or non-substituted alkyl group, a substituted or non-substituted aryl group, or a substituted or non-substituted heterocyclic group; and R⁶ represents a substituted or non-substituted alkyl group having at least 3 carbon atoms, or a substituted or non-substituted aryl group, the method comprising the steps of:

adding a chloroformic acid derivative represented by the following general formula (7) to an isothiocyanic acid salt represented by the following general formula (5) and a hydroxy derivative represented by the following general formula (6) for preparing a first intermediate represented by the following general formula (8);

preparing a second intermediate represented by the following general formula (10) from the first intermediate; and

reacting the second intermediate with an alkylating agent represented by one of the following general formula (11) and the following general formula (12) for preparing the isothiocyanatoformic acid ester derivative:

9

U.S. Application No.: 10/074,014

Attorney Docket No.: Q68466

$$\begin{pmatrix}
0 \\
R^4OCN = C
\end{pmatrix}
C
S$$

$$N$$

General formula (10)

General formula (11)

$$(R^{5}O)_{2}SO_{2}$$

General formula (12)

wherein, in the general formula (5), Z represents a sodium atom or a potassium atom, in the general formula (10), M represents an alkali metal atom, an alkali earth metal atom, an aluminum atom or a magnesium atom, and, in the general formula (11), X represents a halogen atom or SO₃Ar, and Ar represents a substituted or non-substituted aryl group.

U.S. Application No.: 10/074,014

Attorney Docket No.: Q68466

15. (withdrawn): A method of preparing an isothiocyanatoformic acid ester derivative represented by the following general formula (2):

General formula (2)

in which: R⁴ represents a substituted or non-substituted alkyl group, or a substituted or non-substituted aryl group; R⁵ represents a substituted or non-substituted alkyl group, a substituted or non-substituted aryl group, or a substituted or non-substituted heterocyclic group; and R⁶ represents a substituted or non-substituted alkyl group having at least 3 carbon atoms, or a substituted or non-substituted aryl group, the method comprising the steps of:

adding a chloroformic acid derivative represented by the following general formula (7) to an isothiocyanic acid salt represented by the following general formula (5) and a hydroxy derivative represented by the following general formula (6) for preparing a first intermediate represented by the following general formula (8);

preparing a second intermediate represented by the following general formula (10) from the first intermediate; and

reacting the second intermediate with an alkylating agent represented by one of the following general formula (11) and the following general formula (12) for preparing the isothiocyanatoformic acid ester derivative:

11

U.S. Application No.: 10/074,014

General formula (5)

Reformula (6)

General formula (6)

Reformula (6)

General formula (6)

General formula (7)

General formula (8) $R^{4}OCNHCOR^{6}$ General formula (8) $R^{4}OCN = COR^{6}$ General formula (10) $R^{5}X$ $R^{6}OH$ General formula (6)

Attorney Docket No.: Q68466

wherein, in the general formula (5), Z represents a sodium atom or a potassium atom, in the general formulas (9) and (10), M represents an alkali metal atom, an alkali earth metal atom, an aluminum atom or a magnesium atom, and, in the general formula (11), X represents a halogen atom or SO₃Ar, and Ar represents a substituted or non-substituted aryl group.

16. (new): The method of claim 12, wherein in preparing the first intermediate represented by formula (8), the intermediate represented by the formula (8) is prepared by

U.S. Application No.: 10/074,014

Attorney Docket No.: Q68466

dissolving in a reaction solvent the isothiocyanic acid salt represented by the formula (5) and the hydroxy derivative represented by the formula (6), and then adding dropwise the chloroformic acid derivative represented by the formula (7), thus causing a reaction.

17. (new): The method of claim 12, wherein in preparing the first intermediate represented by the formula (8), an amount (mol) of the isothiocyanic acid salt represented by the formula (5) is 1 to 5 times an amount of the hydroxy derivative represented by the formula (6).

18. (new): The method of claim 12, wherein in preparing the first intermediate represented by the formula (8), an amount (mol) of the isothiocyanic acid salt represented by the formula (5) is 1 to 3 times an amount of the hydroxy derivative represented by the formula (6).

19. (new): The method of claim 12, wherein in preparing the first intermediate represented by the formula (8), an amount (mol) of the chloroformic acid derivative represented by the formula (7) is 0.5 to 3 times an amount of the hydroxy derivative represented by the formula (6).

20. (new): The method of claim 12, wherein in preparing the first intermediate represented by the formula (8), an amount (mol) of the chloroformic acid derivative represented by the formula (7) is 0.5 to 1.5 times an amount of the hydroxy derivative represented by the formula (6).

21. (new): The method of claim 12, wherein in preparing the first intermediate represented by the formula (8), a solvent selected from the group consisting of acetonitrile, acetone, and tetrahydrofuran is used as a reaction solvent.

U.S. Application No.: 10/074,014

Attorney Docket No.: Q68466

22. (new): The method of claim 12, wherein preparing the first intermediate represented by the formula (8), a reaction temperature thereof is in a range from -25 to 40°C.

23. (new): The method of claim 12, wherein in preparing the second intermediate represented by the formula (10), an amount (mol) of the compound represented by the formula (9) is 1 to 5 times an amount of the first intermediate represented by the formula (8).

24. (new): The method of claim 12, wherein in preparing the second intermediate represented by the formula (10), a solvent selected from the group consisting of water, methanol, ethanol, acetonitrile, acetone, and tetrahydrofuran is used as a reaction solvent.

25. (new): The method of claim 12, wherein in reacting the second intermediate represented by the formula (10) with the alkylating agent represented by one of the formulas (11) and (12), suspending the intermediate represented by the formula (10) in a reaction solvent and then adding dropwise the alkylating agent represented by the formulas (11) or (12) are included in order to obtain the isothiocyanatoformic acid ester represented by the formula (2).

26. (new): The method of claim 12, wherein in reacting the second intermediate represented by the formula (10) with the alkylating agent represented by one of the formulas (11) and (12), an amount (mol) of at least one of the alkylating agent represented by one of the formulas (11) and (12) is 1 to 3 times an amount of the second intermediate represented by the formula (10).

27. (new): The method of claim 12, wherein in reacting the second intermediate represented by the formula (10) with the alkylating agent represented by one of the formulas (11) and (12), an amount (mol) of at least one of the alkylating agent represented by one of the

Attorney Docket No.: Q68466

formulas (11) and (12) is 1 to 1.5 times an amount of the second intermediate represented by the formula (10).

28. (new): The method of claim 12, wherein in reacting the second intermediate represented by the formula (10) with the alkylating agent represented by one of the formulas (11) and (12), a solvent selected from the group consisting of acetonitrile, acetone, dimethylformamide, dimethylacetylamide, tetrahydrofuran, dimethyl sulfoxide, and alcohol is used as a reaction solvent.